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May 28, 2002

Dr. Michael A. Meyer
NASA HQ 300 E ST SW Code SR
Washington, DC 20546

RE: **Final Technical Report on NAG5-8822**
"Spectroscopic Studies of Pre-Biotic Carbon Chemistry"

Dear Dr. Meyer:

Enclosed please find the Final Technical Report (original and 2 copies) prepared by Professor Geoffrey A. Blake on the above referenced grant.

If you have any questions, please feel free to call me at (626) 395-6481.

Sincerely



Leticia Calderon
Office Assistant

Enclosures

cc: ONRRR/San Diego
NASA Center for Aero Space Information (CASI) ✓
Cheryl D. Lee, Grant Negotiator
Dr. Richard P. Seligman, OSR, CIT
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Grain / N / 90

I. Final Report for Exobiology FY00,01 Funding

As described in the original proposal and in our progress reports for NAG5-8822, research in the Blake group supported by the Exobiology program seeks to understand the pre-biotic chemistry of carbon along with that of other first- and second-row elements from the earliest stages of star formation through the development of planetary systems. The major tool used is spectroscopy, and the program has observational, laboratory, and theoretical components. The observational and theoretical programs are concerned primarily with a quantitative assessment of the chemical budgets of the biogenic elements in star-forming molecular cloud cores, while the laboratory work is focused on the complex species that characterize the pre-biotic chemistry of carbon. We outline below our results over the past two years acquired, in part, with Exobiology support.

Observational Characterizations of Pre-Biotic Chemistry

We continue to employ both Caltech and international facilities in our telescopic studies of pre-biotic chemistry in the circumstellar accretion disks around young stars and in the comae of comets. This part of the Exobiology effort combines observations of young stellar systems at the best spatial resolution achievable, detailed radiative transfer and chemical modeling, and laboratory/spacecraft measurements of grain/grain mantle properties to study the chemical and physical attributes of protostellar, and potentially protoplanetary, nebulae. We are presently following up the results outlined below by extending them to new sources, and on bringing the results to publishable form as part of Jacqueline Kessler's Ph.D. thesis.

During this grant we have completed a number of high resolution studies of the morphology and chemistry of individual YSOs. Students supported by our current and previous Exobiology grants have been involved in making some of the first sub-arcsecond resolution observations with the OVRO array, in continuing our chemical studies of individual systems, in the analysis of mid- and far-infrared spectra taken by ISO, and in continuing exploratory near- and mid-infrared diffraction limited imaging and spectroscopy at the Keck telescopes.

Notable recent accomplishments, described in more detail below, include:

- ◊ optimizing detailed radiative transfer modeling of the molecular and dust emission from YSO envelopes and from comets, including a new parallelized implementation,
- ◊ imaging chemical zonation in the outer regions of T Tauri star accretion disks, particularly fractional ionization and D/H studies,
- ◊ acquiring the first measurements of the CO $v = 1 \rightarrow 0$ emission from the terrestrial planet-forming region of circumstellar accretion disks.

Chemistry in Disks and the Connections to Comets

To date, most spectral line imaging of disks around T Tauri (TTs) and Herbig Ae (HAe) stars has been carried out in isotopomers of CO for reasons of sensitivity (e.g. Dutrey et al. 1994; Koerner & Sargent 1995; Mannings & Sargent 1997). With support from this program we have pioneered studies of the chemical properties of circumstellar disks – properties of great importance to Exobiology. For example, we have imaged the emission from more complex species such as HCO⁺, HCN, and H₂CO interferometrically (see also related single dish work by Dutrey et al. 1997, Kastner et al. 1997). These important first data suggest that chemical studies can now be profitably pursued in at least some disks.

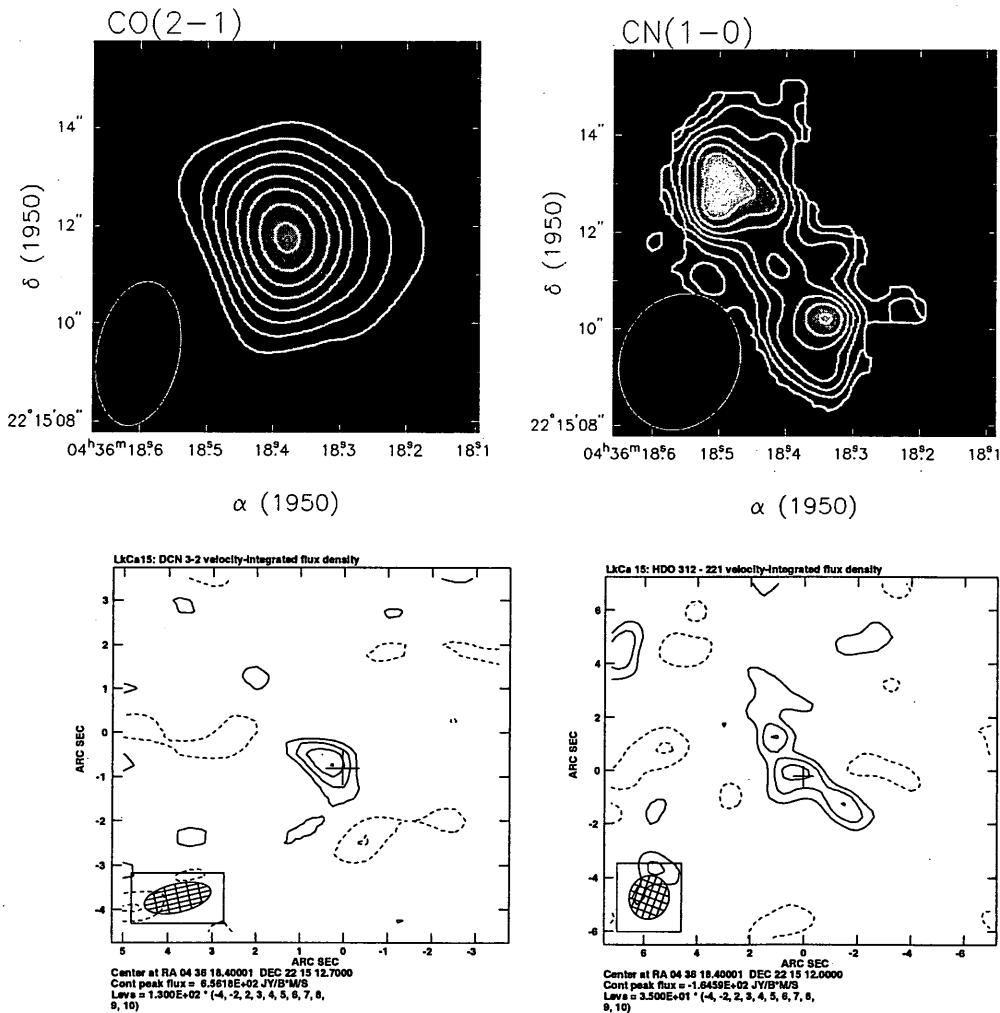


Figure 1. Molecular line emission from the LkCa 15 circumstellar accretion disk (Qi et al. 2002). The top two panels present the integrated emission from the CO and CN, while the lower left and lower right panels depict the emission from DCN and HDO at 1.3 mm.

In order to examine accretion disks in detail and to test models of disk chemistry and transport, we have begun an intensive multi-species imaging study of two T Tauri and two Herbig Ae stars (Qi, Ph.D. Thesis, supported in part by this grant; Qi et al. 2002). The target systems are isolated from molecular clouds and have ages of at least 2 MYr. Interferometric images of several species in the C-, N-, O-, and S-bearing chemical families, including a number of isotopic variants, have been acquired for the first time. Maps of the intense CO and CN emission from the TTs LkCa 15 are shown in Figure 1. Clearly, even at the 3-5'' resolution achieved CO/CN distributions are dramatically different, and reveal enhanced CN abundances outside 200-300 AU. Both ion-molecule and photon-dominated chemistry must contribute to the observed abundances since the CN/HCN and HNC/HCN ratios are too high to be accounted for by quiescent chemistry (Spaans 1996, Dutrey et al. 1997, Kastner et al. 1997), but can be explained by models invoking selective molecular depletion in the outermost regions of the disk (Aikawa & Herbst 1999). The bottom two panels of Figure 1 present the emission from the deuterated isotopomers of HCN and water. Interestingly, the D/H ratio, measured for the first time in the disk of LkCa 15, is very high,

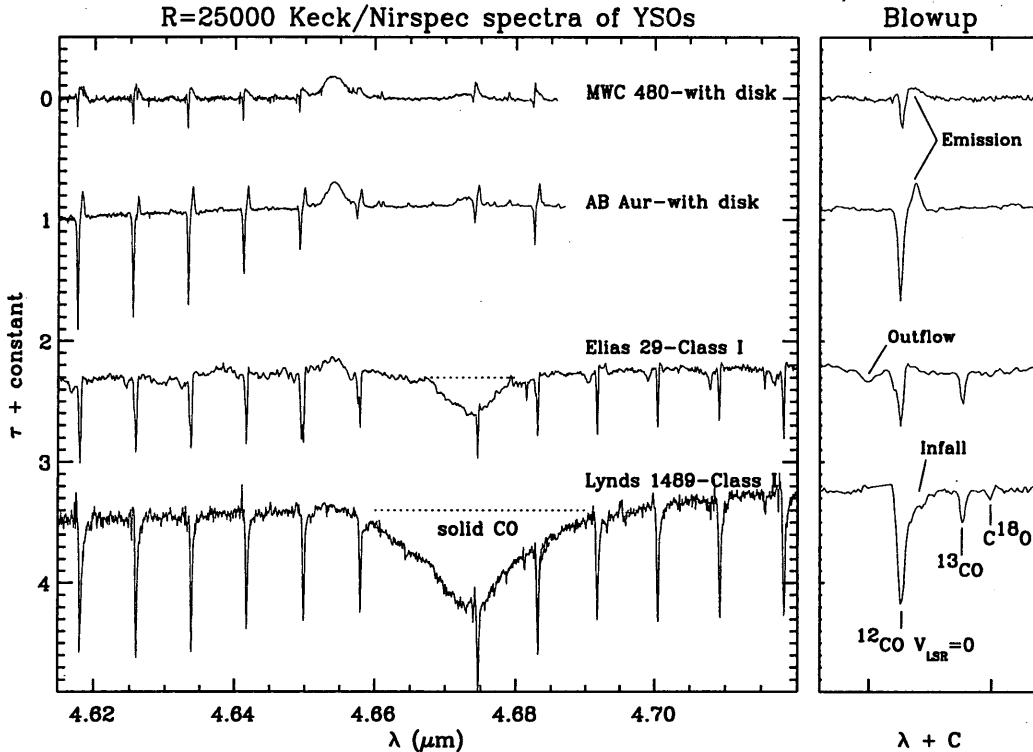


Figure 2. Keck NIRSPEX spectroscopy of the $4.67 \mu\text{m}$ CO fundamental vibration toward low mass protostars at various stages of evolution. Age increases toward the top. The CO ice band visible in the youngest sources with surrounding cores or extended disks (L1489 and Elias 29) is a well established tracer of thermal processing, while the narrow absorption lines directly trace the physical conditions and velocity fields of the surrounding gas. The redshifted wings on the L1489 lines trace infalling gas to within a few stellar radii of the central object (Boogert et al. 2002a). In sources with accretion disks, CO emission becomes visible, and traces warm, dense gas in accretion flows or gaps in the terrestrial planet-forming region. Information on the location of the emitting gas is contained in the line profiles that are clearly resolved with NIRSPEX. The absorption in AB Aur and MWC 480, whose spectra were acquired at high airmasses, is caused by the atmosphere. Measurements at different times of the year can be used to shift the astronomical and atmospheric features and clean up the spectrum (Boogert et al. 2002b).

0.008, and is quite similar to those measured in the cold, dense cores of molecular clouds.

These new measurements of the chemistry in the outer regions of circumstellar disks tell us about the formation and evolution of other protoplanetary nebula, and by inference about our own. On the other hand, comets, among the most pristine remnants of the early solar nebula, provide a historical record of the evolution of ices and gases in our solar system. Millimeter-wave aperture synthesis directly probes the inner coma of comets, a region suffering from obscuration and scattering problems in optical and infrared measurements, but the region containing the most pristine cometary gases. The millimeter-wave arrays can sample small-scale ($\geq 1''$) structures near the nucleus with sub-km s^{-1} velocity resolution.

Our first work in exploring the comet-origins connection was on Comet C/1995 O1 (hereafter Hale-Bopp), an exceptionally large and productive long-period comet that provided an opportunity to study the physical and chemical properties of cometary material in unprecedented detail. At OVRO, emission from twelve different molecules was imaged along with the thermal emission from nucleus and its surrounding coma over a five day period in March/April 1997. The measured (D/H) ratios are similar to those seen in LkCa 15 and further suggest that Hale-Bopp, and by inference the outer solar nebula in general, consists of ~15-40% largely unprocessed interstellar material (Blake et al. 1998). The discrepancy between the comet D/H ratios and that of the earth's oceans likely eliminates long period comets as the major source of water on earth (c.f. Meier et al. 1998).

These millimeter-wave studies are well suited to examining the outermost reaches of circumstellar accretion disks and the outer reaches of the solar system, but tell us little about the terrestrial planet-forming regions of the disks around pre-main sequence stars. The discovery of extrasolar 'hot Jupiters' some 0.05 AU from their parent stars (Marcy, Cochran & Mayor 2000) has highlighted the important role of star-disk-protoplanet interactions, and demands new tools that can investigate the critical 1-10 AU planet-forming zone of disks (Lin et al. 2000, Ward & Hahn 2000, Trilling et al. 1998). The observational characterization of gaps and of the fraction of disks containing Jovian protoplanets therefore forms a pivotal counterpoint to the highly successful radial velocity extrasolar planet searches.

Only high resolution spectroscopy permits robust access to these spatial scales at present (see Najita et al. 2000). Indeed, the potential signatures are very large. Jupiter induces a stellar velocity wobble of only 13 m s^{-1} , for example, while alterations to the disk kinematics can be a large fraction of the orbital velocity of $\sim 20 \text{ km s}^{-1}$ at 5 AU. Gas in and near the $\lesssim 1 \text{ AU}$ gaps opened up by protoplanets can be heated by the star and, if the planet is massive enough, by shocks (Bryden et al. 1999, Kley 1999). The total amount of hot, dense gas is small, but it radiates strongly in the IR since it is adjacent to the low density gap. Abundant species such as H_2 , H_2O , and CH_4 are excellent candidate tracers, but they are very difficult to observe from the ground.

CO is an abundant, stable molecule widely spread throughout the disk (and any envelope, if present). Mm-wave CO emission lines trace nicely the outer disk (Beckwith & Sargent 1996), while $\Delta v = 2$ overtone emission near $2.3 \mu\text{m}$ arises from the several thousand degree gas immediately adjacent to the young star (Scoville et al. 1983, Najita et al. 1996), but can only be observed in a handful of stars. CO absorption at 2.3 and $4.6 \mu\text{m}$ traces cold gas and ice with moderate optical depths, leaving *emission* from the the CO vibrational fundamental near $4.6 \mu\text{m}$ as a potential gap/protoplanet tracer. In a study of 8 spectroscopic binary systems, 5 show measurable CO $v = 1 \rightarrow 0$ emission thought to arise from a gap in the circumbinary disk, an interpretation consistent with the CO line shapes (Najita et al. 2000). For the observed intensities of $\sim 10^{-16} \text{ W m}^{-2}$, the total amount of radiating material is only 10^{-5} earth masses, with a $T \gtrsim 1100 \text{ K}$. For $T \sim 300 \text{ K}$, the same line strength would correspond to a mass of only $\sim 0.01 M_{\oplus}$ at the distance of Taurus!

We have therefore taken a two pronged approach to the study of inner disk regions. Using the ISO satellite we have measured, for the first time, the pure rotational H_2 emission from both accretion and debris disks (Thi et al. 2001a,b). These results have accurately constrained the gas mass in disks for the first time, and have revealed that gas is present around young stars for much longer than previously believed. We have also begun a program

of $4.67\ \mu\text{m}$ CO observations of CO using the NIRSPEC spectrograph at the Keck II telescope. We have studied a carefully selected sample of lines of sight, ranging from quiescent dense clouds, to embedded young protostars, to more evolved, disk dominated protostars. Our initial results demonstrate the tremendous power of this tracer, as outlined in Figure 2.

With continuing Exobiology support we will be mapping YSO's at the highest possible resolution, but with sufficient (u, v) sampling to recover all the emission on scales up to 30-60" using both the OVRO and BIMA millimeter arrays. At 0.2" resolution, gaps or holes similar to those found in debris disks will become observable in dust continuum emission in the nearest star forming regions. Molecular line data are acquired simultaneously with the continuum data. The spectral line data are more difficult to interpret as questions of line optical depth, molecular depletions, continuum optical depth, and vertical structure within the disk all have to be considered. To some extent these complications can be minimized by studying a number of isotopomers. We will model and interpret the molecular line data to elucidate the kinematics and chemistry of the disk and inner envelope. In particular, if the HCN and CN patterns discovered in LkCa 15 do indeed arise from a CO "condensation front", higher resolution observations will be able to measure the location and width of the transition zone directly. Indeed, the degree of volatility of various species controls, in large measure, the distances at which they begin to condense on grain mantles, but this location is very dependent on the disk physics and is difficult to predict theoretically. We also plan to continue our interferometer surveys of long period comets as they appear. A two week OVRO observing session last summer was conducted, for example, on comet S/4 1999 (LINEAR).

The development of state-of-the-art radiative transfer codes continues, as does that for up-to-date gas phase *and* grain mantle chemical codes on platforms at Caltech and Leiden. The chemical codes are well adapted to incorporating dynamical motions of the gas and grain materials. We are using these codes to construct models of the physical and chemical structure of protoplanetary nebulae for those species which are directly observable, and to make testable predictions for those which are not. In this sense, the codes will be chemically similar to those developed elsewhere (Aikawa et al. 1998, Willacy et al. 1998), but will incorporate realistic radiative transfer models for the first time. Much as we have shown to be the case for the (u, v) modeling of high spatial resolution observations of envelope/disk systems, we believe the incorporation of such codes is essential to providing realistic estimates of the observational uncertainties and their impact on the constraint of important physical and chemical processes during star formation.

Our modeling effort is carefully focussed on interpretation of observational data. We believe it will be possible to test qualitatively proposed dynamical models provided the chemical codes adequately include both gas phase and grain mantle components. In all cases, we will concentrate on those species which are known to be present in primitive solar system materials. The models will be used to assess those molecules or materials which are most likely to possess a substantial interstellar component, and to make testable predictions about the abundance and isotopic character of these materials. The calculations are very taxing computationally, and we have implemented a parallel version on the 32 node Beowulf machine in Planetary Science at Caltech. Access to the 320 node machine in Geophysics has been negotiated, and our first detailed 2D and 3D calculations began this spring.

Laboratory Studies of Complex Pre-Biotic Molecules

The Blake group relocated over the past two years to a new laboratory in the Beckman Institute, which provides substantially more space (nearly 2500 sq. feet) and clean room quality air over the optical benches. During the move and the subsequent disruption of our ability to carry out studies under UHV conditions, we have been developing important new light source capabilities. Specifically, we are attempting to improve our studies in two distinct ways: (1) By developing a compact, all solid-state tunable vacuum ultraviolet (VUV) spectrometer, and (2) By extending the range of our mid-IR and far-IR light sources via difference frequency generation techniques in non-linear crystals such as AgGaS_2 and optoelectronic substrates such as low temperature GaAs. By employing a single VUV photon, *any* molecule with an ionization potential less than the photon energy may be studied. At longer wavelengths, difference frequency methods can potentially yield much better output powers and very broad tunability compared with our current spectrometers, including the pivotal THz region. Recent tests of difference frequency mixing in GaAs have revealed cut-off frequencies nearer to 3 THz, which will enable the rotational spectra of *all* interstellar molecules and first-row atomic fine structure transitions to be studied with sensitive SIS receivers from future platforms such as SOFIA and FIRST. Further, the vibration-rotation-torsion spectra of more complex molecules may be best studied at THz frequencies, as outlined below.

The chemistry of carbon is characterized by an extraordinary array of structures and reactivities. In the diffuse interstellar medium this extraordinarily complex chemistry is reflected in our emerging understanding of cosmic dust, in which species as diverse as carbon chains and rings, fullerenes and fullerenes, and polycyclic aromatic hydrocarbons (PAHs) or their derivatives have all been proposed as carriers of the diffuse interstellar bands (DIBs) and unidentified infrared emission features (UIRs) – spectral features which are used to trace the presence of “molecular grains.” In dense interstellar gas, more saturated compounds are rapidly synthesized, and recent theoretical models have suggested that truly pre-biotic species may be formed in considerable abundance (Charnley 2000).

Over the course of the last year we have been examining the millimeter-wave and THz spectra of important pre-biotic molecules with our OPO and difference frequency spectrometers in order to search for such species with the soon to be completed SOFIA and FIRST/Herschel observatories since their softest THz modes may also offer a more sensitive way to search for complex species in dense interstellar clouds. Consider the case of glycine, for example, which has long been sought after unsuccessfully at microwave and millimeter-wave frequencies (Snyder 1997). Even with the nuclear and electronic partition functions set to unity, careful considerations of the glycine vibrational manifold lead to a partition function of $\sim 650,000$ at 300 K! Calculated 0 to 3 THz spectra for the four lowest conformers of glycine (here, conformers refers to the relative orientations of the carboxylic acid and amino functional groups of glycine) show that the THz Q branches are substantially stronger (by factors of 100-1000) than the pure rotational transitions at centimeter and millimeter wavelengths at room temperature. Under low temperature conditions, the relative intensity ratios will be even more in favor of the THz bands.

We have recently installed both heated pulsed nozzles and laser desorption sources, and have created a novel “pick up” source in which two expansions are crossed to create new

species that are too reactive to produce in single expansions. Our first application of this source is described in our work on sodium-water and sodium-ammonia clusters that appeared in *Chemical Physics Letters*. These clusters have low IPs that can easily be accessed with the OPOs, and were used to test the system before moving to pre-biotic species. Important precursors to species such as glycine and alanine are the essentially uncharacterized amino-alcohols (amino-methanol and -ethanol being the smallest examples), and so our work in the coming year will start with these species. We have recently used these instruments to acquire the spectra of aminoethanol, and molecule predicted to be the immediate precursor of the amino acid alanine in hot core chemistry; and dihydroxyacetone, the second simplest sugar after glycoaldehyde, recently detected in hot cores.

II. Publications Since Late 1998 Under the Current Grant

- "Chemical Evolution of Star-Forming Regions" Ewine F. van Dishoeck & Geoffrey A. Blake, *Annual Reviews of Astron. Astrophys.* **36**, pp. 317-68, 1998.
- "Envelope Structure of Deeply Embedded YSOs in the Serpens Molecular Cloud" Hogerheijde, M., van Dishoeck, E., Salverda, J., & Blake, G.A. *ApJ*, **513**, 350, 1999.
- "A Nanosecond Optical Parametric Generator/Amplifier Seeded by an External Cavity Diode Laser" S. Wu, V. Kapinus, & G.A. Blake 1999, *Opt. Commun.* **159**, 74.
- "Molecular Models of Benzene and Selected PAHs in the Gas, Aqueous, and Adsorbed States" James D. Kubicki, Geoffrey A. Blake, & Sabine E. Apitz 1999, *Env. Toxic. Chem.* **18**, 1656.
- "A Traveling-Wave THz Photomixer Based on Angle-Tuned Phase Matching" Shuji Matsuura, Geoffrey A. Blake, Rolf A. Wyss, J.C. Pearson, Christoph Kadow, Andrew W. Jackson, & Arthur C. Gossard 1999, *Appl. Phys. Lett.* **74**, 2872.
- "Sublimation from Icy Jets as a Probe of the Interstellar Volatile Content of Comets" Geoffrey A. Blake, Chunhua Qi, Michiel R. Hogerheijde, Mark A. Gurwell, & Duane O. Muhleman 1999, *Nature* **398**, 213.
- "Envelope Structure of Deeply Embedded Young Stellar Objects in the Serpens Molecular Cloud" Michiel R. Hogerheijde, Ewine F. van Dishoeck, Jante M. Salverda, & Geoffrey A. Blake 1999, *Ap. J.* **513**, 350.
- "The Impact of the Massive Young Star GL 2591 on its Circumstellar Material. I. Temperature and Density Distributions on 100-20,000 AU Scales" Floris van der Tak, Ewine F. van Dishoeck, N.J. Evans, & Geoffrey A. Blake 1999, *Ap. J.* **522**, 991.
- "Submillimeter-Wave Measurements and Analysis of the Ground and $\nu_2 = 1$ States of Water" Pin Chen, J.C. Pearson, Shuji Matsuura, Geoffrey A. Blake, & Herbert M. Pickett 2000, *Ap. J. Suppl.* **128**, 371.
- "Substantial Reservoirs of Molecular Gas in the Debris Disks around Young Stars" Wing-Fai Thi, Geoffrey A. Blake, Ewine F. van Dishoeck, Gerd-Jan van Zadelhoff, J. Horn, E.E. Becklin, V. Mannings, A.I. Sargent, M.E. van den Ancker, & A. Natta 2001, *Nature* **409**, 60.
- "Spectral Energy Distributions of Passive T Tauri and Herbig Ae/Be Disks: Grain Mineralogy, Parameter Dependences, and Comparison with ISO LWS Observations" E.I. Chiang, M.K. Joungh, M.J. Creech-Eakman, C. Qi, J. Kessler, G.A. Blake, & E.F. van Dishoeck 2001, *Ap. J.* **547**, 1077.
- "Chemical Evolution of Protostellar Matter" William D. Langer, Ewine F. van Dishoeck, Edward A. Bergin, Geoffrey A. Blake, Alexander G.G.M. Tielens, Thangasamy Velusamy, & Douglas B. Whittet 2000, *Protostars & Planets IV*, V.G. Mannings, A.P. Boss & S.S. Russell, eds., (Univ. Arizona, Tucson), 29-58.
- "Submillimeter Lines from the Circumstellar Disks around Pre-Main Sequence Stars" Gerd-Jan van Zadelhoff, Ewine F. van Dishoeck, Wing-Fai Thi, & Geoffrey A. Blake 2001, *Astron. Ap.* **377**, 566.

- "Microwave and THz Spectroscopy" Geoffrey A. Blake 2001, *Encyclopedia of Chemical Physics & Physical Chemistry*, J. Moore, N. Spencer, eds. (Institute of Physics Publ., Bristol), pp. 31-44.
- "Unravelling the Chemical Structure of Young Stellar Objects with ALMA" Ewine F. van Dishoeck & Geoffrey A. Blake 2000, *Science with the Atacama Large Millimeter Array*, A. Wootten, ed. (ASP Conference Series, Vol. 235), pp. 89-98.
- "ISO LWS Spectra of T Tauri and Herbig AeBe Stars in Taurus and Ophiuchus" M.J. Creech-Eakman, E.I. Chiang, Ewine F. van Dishoeck, & Geoffrey A. Blake 2002, *Astron. Ap.*, 385, 546.
- "High Resolution 4.7 μm Keck/NIRSPEC Spectra of Protostars. I: Ices and Infalling Gas in the Disk of L1489 IRS" A.C.A. Boogert, M.R. Hogerheijde, & Geoffrey A. Blake 2002, *Ap. J.* **568**, 761.

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